The Effects of Alloying on the Magnetic Phases of EuSe

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§1. Introduction

The magnetic properties of Eu chalcogenides with the NaCl structure are usually described by the first and second nearest neighbor exchange interactions J_1 and J_2. The ferromagnetic J_1 is a strong function of the Eu-Eu distance, while J_2 is less sensitive to it[1,2]. EuSe shows various magnetic phases such as NNSS ($4.6\sim 2.8$ K), NNS($2.8\sim 1.8$ K) and NSNS types($\lesssim 1.8$ K)[3]. (Here, NSNS means that the spin directions within alternative (111) planes are north and south.) The complex magnetic phases of EuSe have been primarily attribued to $J_1 \simeq -J_2$[1], but have not yet been fully explained.

In a previous work[4] the effects of S,Te and Sr substitutions on the magnetic phases of EuSe were investigated by measuring NMR and AC susceptibilities χ_{ac}. In $EuSe_{1-x}Te_x$ and $Eu_{1-x}Sr_xSe$, where the lattice constant a increases with x, the NSNS phase is stabilized as shown in Fig.1. In $EuSe_{1-x}S_x$ the NNSS and ferromagnetic phases exist for $x \lesssim 0.1$ and $x \gtrsim 0.1$, respectively. In this work we have reexamined the magnetic phases of $EuSe_{1-x}S_x$ near $x \approx 0.10$ and have investigated the effect of alloying with nonmagnetic compounds such as SrS,CaS and SmSe by NMR and χ_{ac}.

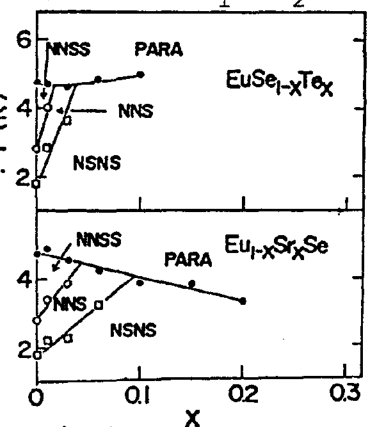

Fig.1

§2. Experimental Results

Polycrystalline specimens were prepared by heating a pressed mixture of powders of EuSe and appropriate compound in an enclosed tantalum crucible at 1700°C for ten hours. The lattice constants of the specimens, as shown in Figs. 2 and 3, decrease with increasing x, following Vegard's law, except for $Eu_{1-x}Sm_xSe$, where a exhibits an anomalous dependence on x owing to a valence change of Sm ions. With increasing x θ_p increases in $EuSe_{1-x}S_x$, while it decreases in other alloys. The x dependences of θ_p are explained on the basis of the molecular field theory.

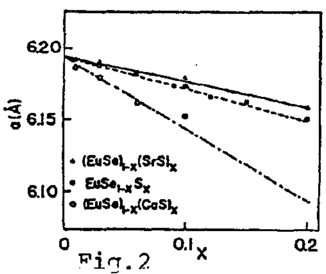

Fig.2

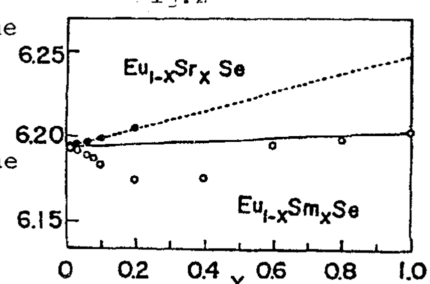

Fig.3

The line shape of the ^{153}Eu NMR in EuSe at 1.7 K consists of two peaks as shown in Fig.4. The high-frequency NMR comes from nuclei on the N sites of the NNS phase(N<u>N</u>S) and the low-frequency NMR comes from the nuclei in the NSNS phase and on the S sites of the NNS phase(NN<u>S</u>)[5]. The NMR of the NNSS phase, the frequency of which agrees with that of the N<u>N</u>S line, is observed above 2.5 K[5]. The difference in the frequencies of the two NMR is that in the transferred hyperfine fields at the two types of Eu sits. χ_{ac} of EuSe, as is shown in Fig.5, exhibits three abrupt changes at 2.0, 2.8 and 4.6 K, which correspond to the NSNS -NNS, NNS-NNSS and NNSS-paramagnetic transitions, respectively.

(1) $EuSe_{1-x}S_x$

The specimens of x = 0.08, 0.09, 0.12 and 0.15 were prepared and investigated. The results, together with previous ones, are shown in Figs.4 and 5. For $x \lesssim 0.10$ only the high-frequency NMR was observed and is attribued to the NNSS phase, since no NNS line was observed. For $x \gtrsim 0.12$ no NMR was detected in the range of 100-200 MHz, probably because of short values of $T_2 (\lesssim 1 \mu sec)$. χ_{ac} for $x \lesssim 0.06$ show only the change due to the first order transition at 4.6 K, indicating , together with NMR results, that only the NNSS phase exists in these specimens. χ_{ac} for $x \gtrsim 0.15$ show typical ferromagnetic behaviors, indicating the existence of the ferromagnetic order, although the NMR confirmation was unscessful. The specimens of x= 0.08 and 0.09 are in the NNSS phase as suggested by NMR and χ_{ac} results, but show no first order transition at $T_N \simeq 4.0$ K. The specimens with $0.10 \lesssim x \lesssim 0.12$ are in the transition from the NNSS to ferromagnetic phase. The magnetic phase diagram of $EuSe_{1-x}S_x$ is shown in Fig.6.

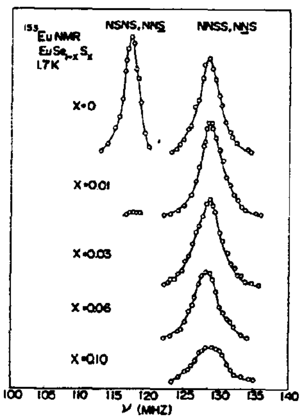

Fig.4

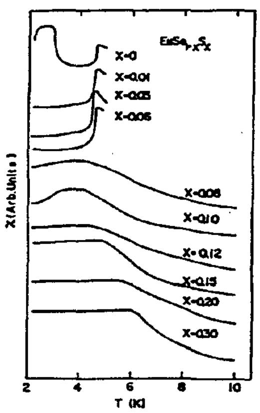

Fig.5

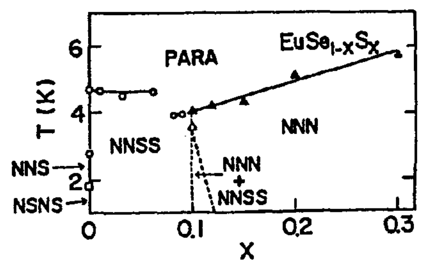

Fig.6

(2) Alloys with SrS, CaS and SmSe

The specimens of $(EuSe)_{1-x}(SrS)_x$ with $x \lesssim 0.2$, $(EuSe)_{1-x}(CaS)_x$ with $x \lesssim 0.1$ and $Eu_{1-x}Sm_xSe$ with $x \lesssim 0.2$ were investigated. Typical results of NMR and χ_{ac} are shown in Figs.7 and 8. Only the high-frequency NMR were observed in $(EuSe)_{1-x}(SrS)_x$ with $x \gtrsim 0.06$ and in $(EuSe)_{1-x}(CaS)_x$ with $x \gtrsim 0.01$. χ_{ac} for $(EuSe)_{1-x}(CaS)_x$ show only the changes at T_N, at which no first order transition occurs for $x \gtrsim 0.06$. These indicate that the NNSS phase is stabilized with increasing x in these alloys as in $EuSe_{1-x}S_x$. But no ferromagnetic order appeared. The results for $Eu_{1-x}Sm_xSe$ are similar, except for the critical concentration of x = 0.10, above which only the NNSS phase exists.

In Fig.9 the Néel temperatures in various alloy systems are plotted against a. The temperatures of the NSNS-NNS and NNS-NNSS transitions are also drawn by

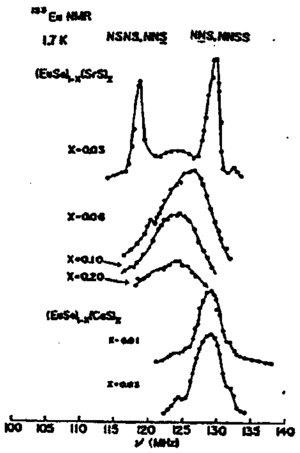

Fig.7

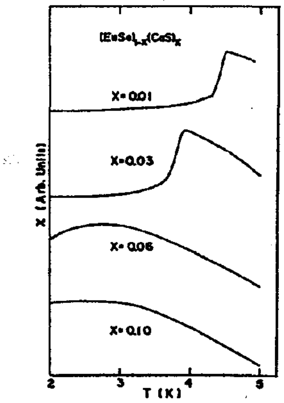

Fig.8

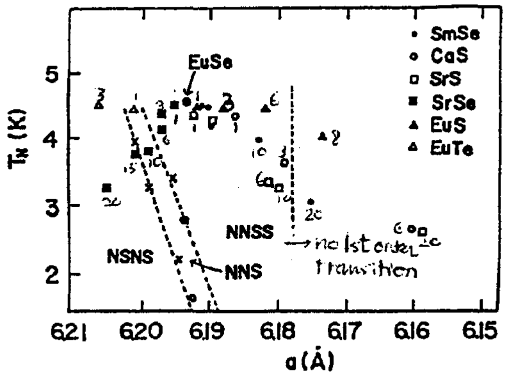

Fig.9

the points(X) and broken curves. The transition temperatures exhibit different dependences on x among different alloy systems, but their dependences on a lie on smooth curves, suggesting that the changes of the magnetic phases in alloying are primarily induced by the changes in a. As seen in Fig.9, the NSNS and NNSS phases are stabilized with increasing and decreasing a, respectively, and the NNS phase is stable in a narrow range of a. The first order transition at T_N disappears below a = 6.175~1.180 A.

§3. Discussions

In the molecular field approximation using J_1 and J_2 the magnetic phase is determined by J_2/J_1[1]. Various phase changes of EuSe in alloying may be primarily due to $J_2/J_1 \simeq -1$ and a strong dependence of J_1 on a. The NSNS phase in $EuSe_{1-x}Te_x$ and $Eu_{1-x}Sr_xSe$ and the ferromagnetic phase in $EuSe_{1-x}S_x$ are qualitatively understood by a decrease and an increase of J_1, respectively, which are caused by the changes of a in alloying. But the NNSS and NNS phases, including the first order transition at T_N, cannot be explained only in terms of J_1 and J_2. Other mechanisms such as lattice distortion[6] and higher order exchanges[7] might play important role in determining the magnetic phases.

References

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